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Newly developed multilayer-based mirrors and optical elements enable the imaging of high-resolution structure and ultrafast dynamics of samples with the soft X-ray free electron laser, FLASH, at DESY in Hamburg.

The FLASH free-electron laser (FEL) produces intense ultrashort soft X-ray pulses with more than 10^8 times higher peak brightness as compared with the most advanced synchrotron radiation sources. This allows time-resolved X-ray imaging and holography of nanostructures with a temporal resolution approaching 10 fs, opening up new studies of laser-matter interactions and the dynamics of correlated systems. In addition, the ultrafast pulses can be used to obtain structural data before the onset of radiation damage. This vastly increases the dose that can be used to record images of biological samples and hence improving the resolution of images.

The extreme power of the X-ray pulses poses a challenge, and new methods are required to harness them. The methods developed here will also pave the way to imaging at upcoming hard-X-ray FELs. With those sources, atomic-resolution imaging of single uncrystallized macromolecules may become possible.

In the first demonstration of ultrafast X-ray imaging at FLASH, a micron-sized test object was illuminated by a single focused coherent FEL pulse[1] (Fig. 1). The coherent diffraction pattern of the object was recorded in the far field on a CCD detector. This pattern was numerically transformed to a high-resolution image of the object, using an iterative phase retrieval technique.[2] This image, formed without the use of a lens, has a resolution limited only by the wavelength and the angular extent of the CCD detector. The lensless nature of coherent diffractive imaging has the advantage that no optical element need be placed near the object, and it is not necessary to carefully position the object—focusing is performed numerically in the phase retrieval step.[3] However, the experiments at FLASH depended critically on ability to measure the forward scattering from samples with high sensitivity and low contamination.

The main experimental challenges are posed by the high pulse intensities, which can reach 10^{15} W/cm^2 in our experiments. We must prevent the direct (undiffracted) FEL beam from hitting and destroying the direct-detection CCD and to prevent out-of-band radiation (plasma emission from the sample) or non-sample scatter from obscuring the coherent diffraction signals. We solved these problems with a unique design that consists of a flat mirror oriented at 45° to the beam with a hole in the middle (Fig. 2). The direct FEL beam passes through a hole in the mirror whereas the diffracted beam is reflected from the mirror onto a bare CCD. Our mirror design enabled the camera to record diffraction angles between –15° to +15°. To reflect scattered light over this wide angle range required a multilayer coating with a very steep lateral gradient. Indeed, the multilayer design had to double in period over only 28 mm. Coherent diffractive imaging was performed with cameras operating at 32 nm, 16 nm, 13.5 nm and 4.5 nm, each utilizing a different multilayer design.[4] The shorter the wavelength the narrower the reflectivity peak width and the higher the specifications for wavelength matching across the optic.

Multilayer coatings are artificial structures that may be designed to enhance reflectivity through constructive interference of beams reflected from the many layer interfaces. Such structures are necessary to efficiently reflect soft X-rays at angles of incidence steeper than the critical angle. Lawrence Livermore National Laboratory (LLNL) is a leader in design and fabrication of multilayer x-ray optical components (including lenses, mirrors, beam splitters, synthetic holographic optical components) for the last 25 years.[5] These capabilities were a key ingredient in the success of the Extreme Ultraviolet Lithography project carried out at LLNL and other laboratories.[6] Multilayers with nanometer periods are now indispensable in cutting-edge experiments at FELs, not only as X-ray optics but also as samples.

One such application was the use of a multilayer film as a nanostructured sample to study the interaction of FEL pulses with matter. In this case, the measurement of the multilayer reflectivity provided a very accurate way to monitor changes in the atomic positions and the refractive indices of the materials in the layers. In experiments at FLASH it was demonstrated that no structural damage occurred within the multilayer during the short (25 fs duration) FEL pulse to within 0.3 nm.[7]

The coherent diffractive imaging technique, with its simple experimental design, has produced perhaps the fastest images ever taken.[8] With future developments in FEL optics it will be possible to perform time-resolved imaging of processes induced by pulses of the same or different wavelength, synchronized to the imaging (probe) pulse. In this way it will be possible to study ultrafast phase transitions, or dynamic effects such as crack or shock propagation, with nanometer spatial resolution and femtosecond temporal resolution.

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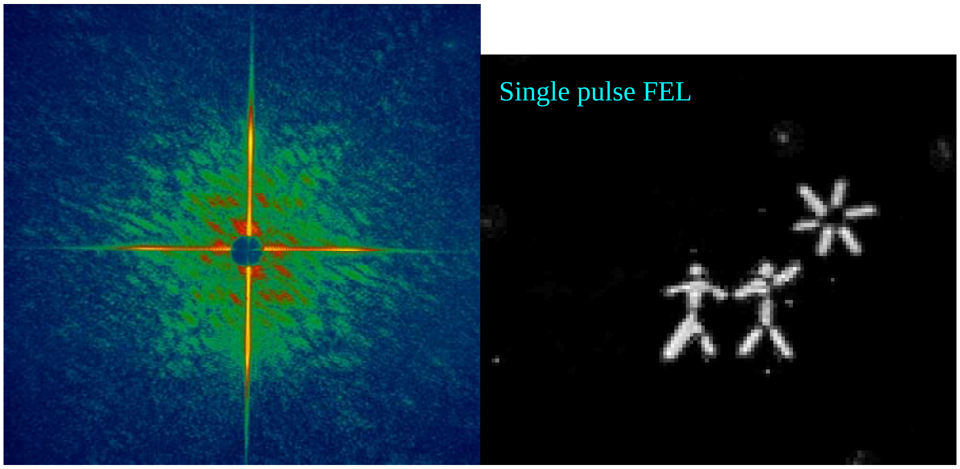

Fig. 1: (Left) A coherent diffraction pattern recorded with a single FEL pulse of 32 nm wavelength from a microfabricated test object. (Right) The image reconstructed from the diffraction pattern by phase retrieval. The lateral extent of the image is 7.5 micron. The resolution of the image is 62 nm.

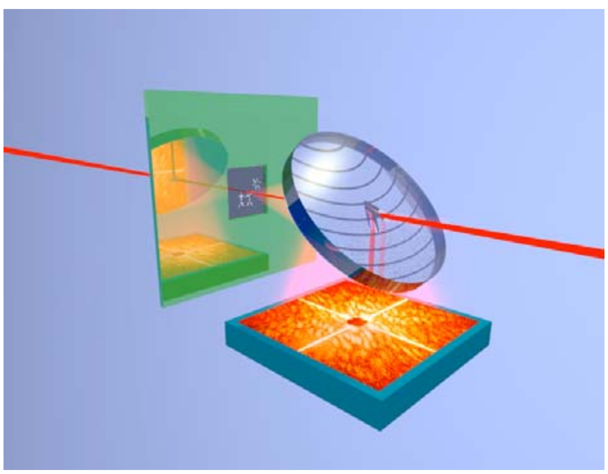

Fig. 2: The concept of lensless coherent diffractive imaging: Multilayer-coated mirror reflects the diffraction pattern onto a CCD, while the direct FEL beam (red) goes through a hole unaffected.